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A FRESH LOOK AT THE CLASSICAL APPROACH TO
HOMOGENEOUS SOLID PROPELLANT
COMBUSTION MODELING

Martin S. Miller
Terence P. Coffee

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US ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND
BALLISTIC RESEARCH LABORATORY
ABERDEEN PROVING GROUND, MARYLAND

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Abstract (Cont'd):

20. broad trends is of greatest heuristic interest, five different propellant data sets are developed which span a wide range of kinetics values. Accurate calculations of the burning rate pressure and temperature dependence are made and parallels drawn with actual propellant behavior. Likely weaknesses in the basic idealization are then identified.

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I. INTRODUCTION

In the past 40 years considerable effort has been directed toward analyzing the physical and chemical processes involved in solid propellant combustion. Part of this effort has been concerned with the development of idealized models of the combustion process as a whole in order to determine how the magnitude, speed, and site of the chemical energy release controls the burning rate. With few exceptions a common thread running through these treatments is the use of the experimental burning rate or its derivatives (i.e., the pressure and temperature sensitivities) as the authenticating test of the model. Because of its long chain of continuity and its comparative dominance of the field we shall term this validation scheme the Classical Approach. In this paper we shall reexamine the utility and limitations of this approach in unfolding solid propellant burning mechanisms.

For the sake of clarity our usage of a few key terms should be explained. We consider that a solid propellant combustion model possesses two essential features: an idealization of the burning process, i.e., a set of differential equations and boundary conditions presumed to express formally the combustion mechanism, and an algorithm or computational scheme for solving these equations. The word "mechanism" is used to describe the collection of fundamental physical and chemical processes proposed as responsible for the self-sustained burning.

The most commonly assumed mechanism, and the one to which we shall confine our analysis here, consists of an exothermic surface pyrolysis reaction which provides the reactants for a single exothermic gas phase reaction. One formal idealization corresponding to this mechanism, developed in detail in Reference 1 involves such assumptions as Arrhenius reaction rates, constant specific heats and thermal conductivity, and unity Lewis number. These assumptions are typical of models addressing the above mechanism. The source of greatest variety among the various models is the computational algorithm, which invariably involves further simplification of the idealization.

The Classical Approach to propellant modeling is most often implemented by developing a model and then varying the kinetics parameters until the model burning rates closely match the experimental burning rates. Apart from limitations in the Classical Approach itself (to be discussed) this particular method of implementation raises two objections. The first is that our perception of the phenomenology arising from the assumed mechanism depends on the fidelity of the model algorithms, and these, for the most part, have not been tested. Thus it is possible (see the NC2 case²) for an algorithm to be particularly inaccurate for the set of parameters deduced from burning rate data using this very algorithm. The question of algorithm reliability is treated in a companion paper.² A second, more subtle objection to this method arises from the lack of uniqueness associated with the determined parameter set. The functional dependence of the experimental burning rate on

¹Miller, M.S., "In Search of an Idealized Model of Homogeneous Solid Propellant Combustion," *Combustion and Flame*, Vol. 46, pp. 51-73, 1982.

²Miller, M.S. and Coffee, T.P., "On the Numerical Accuracy of Solid Propellant Combustion Models," accepted for publication in *Combustion and Flame*.

pressure is usually quite simple, requiring only two or three parameters for a close fit using empirical functions such as $r = a + bP$ or $r = a + bP^n$. Since typically some half dozen model parameters are "floated", it is an open question whether a successful fit is due to the correctness of the assumed mechanism or simply due to the functional flexibility afforded by the large number of disposable parameters. In view of the fact that the mechanism being considered is known (even intended) to be a crude representation of the actual phenomena (in order to isolate the dominant factors), we believe that an exact match of model to experiment is simply not a meaningful objective.

Recognizing that the rationale for propellant modeling is essentially heuristic, one might be better advised to explore thoroughly the range of burning rate behavior implicit to the basic idealization. With such a broad understanding of the model phenomenology one could then attempt to identify similar trends in real propellant combustion. Such a strategy is clearly incapable of validating the proposed mechanism; however, we contend that this limitation is intrinsic to the Classical Approach itself. Further progress in the determination of the physical mechanisms, in our opinion, can only result from more detailed experimental constraints on the structure of the combustion wave. The Classical Approach is no more and no less than a preliminary test of the internal consistency and compatibility of potentially useful concepts.

The central task of this paper then is to explore the range of behavior of the burning rate expected of the stated idealization using a numerically accurate algorithm. This undertaking is complicated by the possibility that the burning phenomenology may depend on the values assigned to the kinetics parameters. To overcome this obstacle we have put together a number of different data sets which cover a wide range of parameter combinations. We believe that the range is sufficiently great to reveal thoroughly the behavior patterns inherent to the mechanism. One can think of these data sets coupled with the idealization as model propellants. Following the Classical Approach we shall compare the way these model propellants burn with real propellants and attempt to draw parallels.

II. THE MODEL PROPELLANTS

Five data sets will be developed in this section by several different methods. RDX1 and NC1, suggestive of nitramine and nitrate ester propellants, are obtained by making plausible assumptions and estimates based largely on experiments not involving steady-state burning. NC2 is based on the kinetics inferred by the method of fitting a model to double-base rate data. The self-consistency of this procedure is examined elsewhere;² we use these parameters here simply to explore as wide a range of kinetics as possible. NC3 is assembled almost entirely from Zenin's analysis³ of embedded thermocouple data taken under actual burning conditions. T1 was put together as a test case to exploit the availability of an exact analytic solution found for the heat feedback under certain conditions¹. This₂ test case also served as a partial check on the numerical code used in this work². The parameter values determined for each of the model propellants are summarized in Table 1.

³Zenin, A.A., "Formal Kinetic Characteristics of the Reactions Accompanying the Burning of a Powder," *Fizika Goreniya i Vzryva*, Vol. 2, pp. 28-32, 1966.

TABLE I.

| | RDX1 | NC1 | NC2 | NC3 | T1 |
|--------------------------------|--------------------------------|----------------------|---------------------------------|-------------------------------|------------------------------|
| M_O (gm/cm ² sec) | 1.035E12 | 8.0E10 | 3456 | 460 | 1.0E12 |
| E_S (kcal/mole) | 47.8 | 42 | 10 | 8 | 45.0 |
| Q_S (cal/gm) | 160 | 75 | 53 | 75 | 75 |
| A_G | 2.5E16 (sec ⁻¹) | 2.5E9 (cc/gm sec) | 1.44E12 (sec ⁻¹) | 1.2E5 (sec ⁻¹) | 100T (sec ⁻¹) |
| E_G (kcal/mole) | 46.2 | 15 | 54 | 5 | 0 |
| Q_G (cal/gm) | 460 | 400 | 422 | 350 | 500 |
| W_B (gm/mole) | 222 | 40 | 40 | 40 | 50 |
| ν | 1 | 2 | 1 | 1 | 1 |
| m_B^{-0} | .48 | 1 | 1 | 1 | 1 |
| N_2 | 6 | 1.33 | 1.33 | 1.33 | 1 |
| ρ_S (gm/cm ³) | 1.6 | 1.6 | 1.55 | 1.6 | 1.6 |

$$\lambda = 2.0E - 4 \text{ cal/cm sec}^\circ\text{K}$$

$$C_p = .35 \text{ cal/gm}^\circ\text{K}$$

A. NC1

The rate of slow thermal decomposition of nitrocellulose (NC) and NC-based propellants has been measured by a number of methods including weight loss⁴, infrared signature⁴, evolution of gas in solvents⁵, and pulsed calorimetry.⁶ Expressed in Arrhenius form the frequency factors range from 10^{18} to 10^{19} sec⁻¹ and the activation energies from 42 kcal to 46 kcal. Thompson and Suh⁷ used an ignition delay technique and obtained a frequency factor of 10^{17} sec⁻¹ with an activation energy of 40 kcal/mole. As representative of these measurements we take the frequency factor (A_s) to be 7.5×10^{17} sec⁻¹ and the activation energy (E_s) to be 42 kcal/mole.⁵ This rate is used for the controlling solid reaction and is assumed to occur only at the surface of the burning propellant. Since this activation energy is suggestive of the endothermic scission of NO₂, we shall assume that subsequent exothermic polymer breakdown reactions occur quickly on the surface leading to a net exothermic heat release Q_s . The embedded thermocouple measurements of Kubota, et.al.⁸ and Zenin⁹ indicate a value of Q_s dependent on the burning rate in the range 60-130 cal/gm. We assume the constant value 75 cal/gm. In order to obtain a surface pyrolysis law in the customary form

$$M = M_0 \exp(-E_s/RT_s) \quad (1)$$

M_0 is constructed as the product of the frequency factor, the number of molecules per unit surface area, and the mass per molecule (W_A).

$$M_0 = \left(\frac{\rho_s^2 W_A}{N_0} \right)^{1/3} A_s \quad (2)$$

The propellant mass density is ρ_s and N_0 is Avogadro's number.

⁴Phillips, R.W., Orlick, C.A. and Steinberger, R., "The Kinetics of the Thermal Decomposition of Nitrocellulose," *J. Phys. Chem.*, Vol. 59, p. 1034-1039, 1955.

⁵Smith, R.D., "Pyrolysis of Dissolved Nitrocellulose," *Nature*, Vol. 170, p. 844-845, 1952.

⁶Aleksandrov, V.V., Bufetov, N.S., Pastukhova, T.V. and Tukhtaev, R.K., "Use of Pulsed Calorimetry for Investigating the Kinetics of Reactions in Condensed Media," *Fizika Goreniya i Vzryva*, Vol. 9, pp. 75-83, 1973.

⁷Thompson, C.L. and Suh, N.P., "The Interaction of Thermal Radiation and M-2 Double-Base Solid Propellant," *Comb. Sci. Tech.*, Vol. 2, pp. 59-66, 1970.

⁸Kubota, N., Ohlemiller, T.J., Caveny, L.H. and Summerfield, M., "The Mechanism of Super-Rate Burning of Catalyzed Double-Base Propellants," AMS Report No. 1087, Princeton University, March 1973.

⁹Zenin, A.A., "Structure of Temperature Distribution in Steady-State Burning of a Ballistic Powder," *Fizika Goreniya i Vzryva*, Vol. 2 pp. 67-76, 1966.

There is evidence¹⁰ that aldehyde-NO₂ reactions may play a significant role in the fizz zone energy release. In the NCl data set we assume they are dominant. Although the rates of these reactions are not precisely known, we take a frequency factor of $2.5 \times 10^9 \frac{\text{cc}}{\text{gm sec}}$ and an activation energy of 15 kcal/mole to be representative.¹¹ The values of other necessary parameters in Table 1 are reasonably consistent with this assumed reaction identity although the nascent reactant mass fraction (mg^{-0}) is assigned arbitrarily. Values for the thermal conductivity and specific heat are assumed to be the same for all of the model propellants and are estimated from Zenin.⁹

B. RDX1

The most comprehensive attempt to model RDX monopropellant combustion is due to BenReuven, et al.^{12,13,14} This model assumes that the surface regresses primarily by RDX evaporation from a melt layer. Two gas-phase reactions are then considered; unimolecular decomposition of RDX vapor and formaldehyde-NO₂ reactions. The latter are found to be responsible for only a small percentage (~15% at 40atm) of the heat feedback. Although exothermic decomposition of RDX is allowed in the melt layer, it appears that the net effect of these processes is endothermic. Very recent embedded thermocouple results¹⁵ for RDX composite propellants (~80% RDX), however, indicate an even higher exothermicity to the surface reaction than for NC-based propellant.

Using these observations to simplify the combustion mechanism to only two reactions, we have developed the following conjectural picture of RDX combustion. When an RDX molecule at or very near the surface decomposes, the chemical energy which becomes available may be used in either of two ways. One or more unreacted

¹⁰Lengelle, G., Duterque, J., Verdier, C., Bizot, A. and Trubert, J., "Combustion Mechanisms of Double Base Solid Propellants," *Seventeenth Symposium (International) on Combustion*, The Combustion Institute, pp. 1443-1451, 1978.

¹¹Pollard, F.H. and Wyatt, R.M.H., "Reactions Between Formaldehyde and Nitrogen Dioxide I.," *Trans. Faraday Soc.*, Vol. 45, pp. 760-767, 1949.

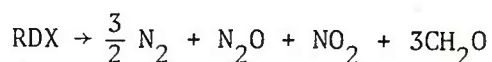
¹²BenReuven, M., Caveny, L.J., Vichnevetsky, R.J., Summerfield, M., "Flame Zone and Sub-Surface Reaction Model for Deflagrating RDX," *Sixteenth Symposium (International) on Combustion*, The Combustion Institute, pp. 1223-1233, 1977.

¹³BenReuven, M., and Caveny, L.H., "Nitramine Flame Chemistry and Deflagration Interpreted in Terms of a Flame Model," *AIAA Paper 79-1133*, AIAA/SAE/ASME 15th Joint Propulsion Conference (Las Vegas, NV), June 18-20, 1979.

¹⁴BenReuven, M., and Caveny, L.H., "Nitramine Monopellant Deflagration and General Nonsteady Reacting Rocket Chamber Flows," *MAE Report No. 1455*, Princeton, University, January 1980.

¹⁵Kubota, N., "Combustion Mechanisms of Nitramine Composite Propellants," *Eighteenth Symposium (International) on Combustion*, The Combustion Institute, pp. 187-194, 1981.

molecules may use this energy to escape the surface bonding forces into the gas phase, or the energy may be absorbed into the surface as heat. In the absence of more detailed knowledge we assume a simple statistical partitioning of the energy among these possible modes. If we assume with BenReuven, et al¹² that RDX decomposes into the following intermediates



then the enthalpy change (solid RDX to gaseous products) at 298°K is 317 cal/gm of RDX. (Heat of formation for RDX is from Dobratz¹⁶ and that for the products from the JANNAF tables¹⁷). Assuming that half of the 317 cal/gm is used to eject unreacted RDX molecules and half goes to surface heating, we obtain $Q_s \doteq 160$ cal/gm. Using the heat of sublimation measured by Cundall¹⁸ (145 cal/gm) as a measure of the surface bonding energy of RDX molecules, we compute that 1.09 gm (=317/2/145) of RDX is ejected unreacted into the gas phase per gram of RDX that undergoes surface pyrolysis. Using Eq. (2) and noting that the surface recedes as a result of RDX loss by pyrolysis and ejection,

$$M_o = (1+1.09) \left(\frac{\rho_s^2 W_A}{N_o} \right)^{1/3} A_s \frac{\text{gm}}{\text{cm}^2 \text{ sec}} \quad (3)$$

The condensed phase kinetics (specifically, the liquid phase) for RDX are taken from the review by Schroeder.¹⁹ To be consistent with this use of liquid phase kinetics, an explicit treatment of the melt layer in our energy partitioning calculation might be desirable; however the above estimates are adequate to the purpose of this paper.

After surface decomposition by what might be termed "dispersive pyrolysis", it is assumed that only the vapor phase decomposition of the unreacted RDX results in significant heat feedback to the surface. This rate is also taken from Schroeder.¹⁹ The gas-phase reaction heat is obtained by summing the heat of sublimation and the reaction enthalpy of solid RDX to gaseous products.

¹⁶Dobratz, B.M., "Properties of Chemical Explosives and Explosive Simulants," University of California (Livermore) Report No. UCRL-51319, Rev. 1, July, 1974.

¹⁷Stull, D.R. and Prophet, H. JANAF Thermochemical Tables, 2nd Edition, NSRDS-NBS-37, June 1971.

¹⁸Cundall, R.B., Palmer, T.F., Wood, C.E.C., "Vapor Pressure Measurements on Some Organic High Explosives," J. Chem. Soc., Faraday Trans., Vol. I, pp. 1339-1345, 1978.

¹⁹Schroeder, M.A., "Critical Analysis of Nitramine Decomposition Data: Activation Energies and Frequency Factors for HMX and RDX Decomposition," Proceedings of 17th JANNAF Combustion Meeting, CPIA Pub. No. 329, Vol. II, pp. 493-508, November 1980.

C. NC 2

Beckstead²⁰ has recently applied the Classical Approach to double base propellants of varying heats of explosion (HEX). In order to achieve a fit of the standard BDP monopropellant model²¹ to the burning rate data at a given HEX, the model "flame" temperature T_f (actually the dark zone temperature) was allowed to vary with pressure. Thus at a given pressure the usual BDP formalism applies. Values of E_s and E_G were determined as 10 kcal/mole and 54 kcal/mole, respectively. Not all of the other fitted parameters were reported but we have used what information was given to construct a consistent data set for HEX=1000 cal/gm at 136 atm, the highest pressure considered. In order to do this we first assumed the values of m_B^{-0} , λ , W_B , and N_2 shown in Table 1.

D. NC3

Zenin^{3,9,22,23} has reported extensive measurements and analysis of temperature distributions in burning double-base propellants obtained by the embedded thermocouple technique. Analysis of the spatial heat release in the condensed phase suggests a reactive zone of at least 10 microns thickness. However, in order to use the Eq. (1) form of the pyrolysis law, we have constructed an Arrhenius plot of M vs. $1/T_s$ and determined values of M_0 and E_s consistent with Zenin's measurements.^{9,23} As in NC1, we have taken Q_s to be 75 cal/gm.

The fizz zone frequency factor was deduced as follows. Values of 1,40, and 30 were assigned to m_B^{-0} , W_B , W_C , respectively. From Reference 9 a value of $Q_G = 350$ cal/gm was selected. Zenin³ determined the fizz zone activation energy to be 5 kcal/mole and the reaction order to be one. We then solve the expression¹

$$q = C_p (T_f - T) \left(\frac{\bar{W}_P}{RT} \right) A_G \exp(-E_G/RT) \quad (4)$$

for A_G at the peak in the fizz zone energy release profile shown in Fig. 1 of Reference 3.

²⁰ Beckstead, M.W., "Model for Double-Base Propellant Combustion," *AIAA Journal*, Vol. 18, pp. 980-985, 1980.

²¹ Beckstead, M.W., Derr, R.L. and Price, C.E., "The Combustion of Solid Monopropellants and Composite Propellants," *Thirteenth Symposium (International) on Combustion*, The Combustion Institute, pp. 1047-1056, 1971.

²² Zenin, A.A., "Burning of Nitroglycerine Powder in Vacuum and at Subatmospheric Pressures," *Fizika Goreniya i Vzryva*, Vol. 2, pp. 74-78, 1966.

²³ Zenin, A.A. and Nefedova, O.I., "Burning of Ballistic Powder Over a Broad Range of Initial Temperatures," *Fizika Goreniya i Vzryva*, Vol. 3, pp. 45-53, 1967.

The test case T1 is assigned values for the pyrolysis law in the same general range as NC1 and RDX1. The temperature dependence of the frequency factor for the gas reaction is chosen so that the reaction rate depends only on the local mass fraction and not on local temperature. This enables use of an exact solution for the heat feedback (Eq. (31) of Ref. 1). The range of values for unimolecular reactions is large^{24,25} and the chosen rate lies well within this range. The remaining parameter values are chosen to be similar to the other data sets.

III. REAL VS. MODEL PROPELLANT COMBUSTION

In this section we shall examine the combustion patterns peculiar to the chosen idealization and determine the extent to which these patterns parallel real propellant combustion. Though many calculations of burning rate based on this idealization have been published we contend that these patterns have not been reliably identified due to the limitations of using a single data set or an inaccurate algorithm or both. Here, with the exception of the NC2 and T1 data sets, the model propellant burning rates were calculated by accurate numerical integrations.² The asymptotic theory of Williams²⁶ modified by the adiabatic deflagration limits of Buckmaster, et al²⁷ was used for NC2 after validation² by numerical integration at 136atm for T_0 in the range -123°C to $+77^\circ\text{C}$. The heat feedback for the T1 case could be obtained analytically (using Eq. 31 of Ref. 1) so that the burning rate determination is simply algebraic (although non-analytic). The variation of these model burning rates with pressure and initial temperature is shown in Figs. 1-10.

The general pressure dependence of the model burning rates is found to be well approximated by the expression $r = a + bP^n$ where n_1 is slightly less than half the gas reaction order (ν). As discussed elsewhere¹ the idealization considered leads to a constant rate at sufficiently low pressure where the gas-phase heat-feedback is negligible compared with the solid-phase heat release. With a laminar gas flame the pressure index is exactly $\nu/2$ so that the solid regression rate has much the same character over at least some pressure range. In general the higher the gas reaction activation energy the closer n is to $\nu/2$. The high

²⁴ Adams, G.F., "An Analysis of the Pressure Dependence of Nitrate Ester Thermal Decomposition," Ballistic Research Laboratory Technical Report ARBRL-TR-02106, September, 1978.

²⁵ Adams, G.F., "A Priori Estimation of Rate Constants for Unimolecular Decomposition Reactions," Ballistic Research Laboratory Technical Report ARBRL-TR-02143, February, 1979.

²⁶ Williams, F.A., "Quasi-Steady Gas-Flame Theory in Unsteady Burning of a Homogeneous Solid Propellant," *AIAA Journal*, Vol. 11, pp. 1328-1330, 1973.

²⁷ Buckmaster, J.D., Kapila, A.K., and Ludford, G.S.S., "Linear Condensate Deflagration for Large Activation Energy," *Acta Astronautica*, Vol. 3, pp. 593-614, 1976.

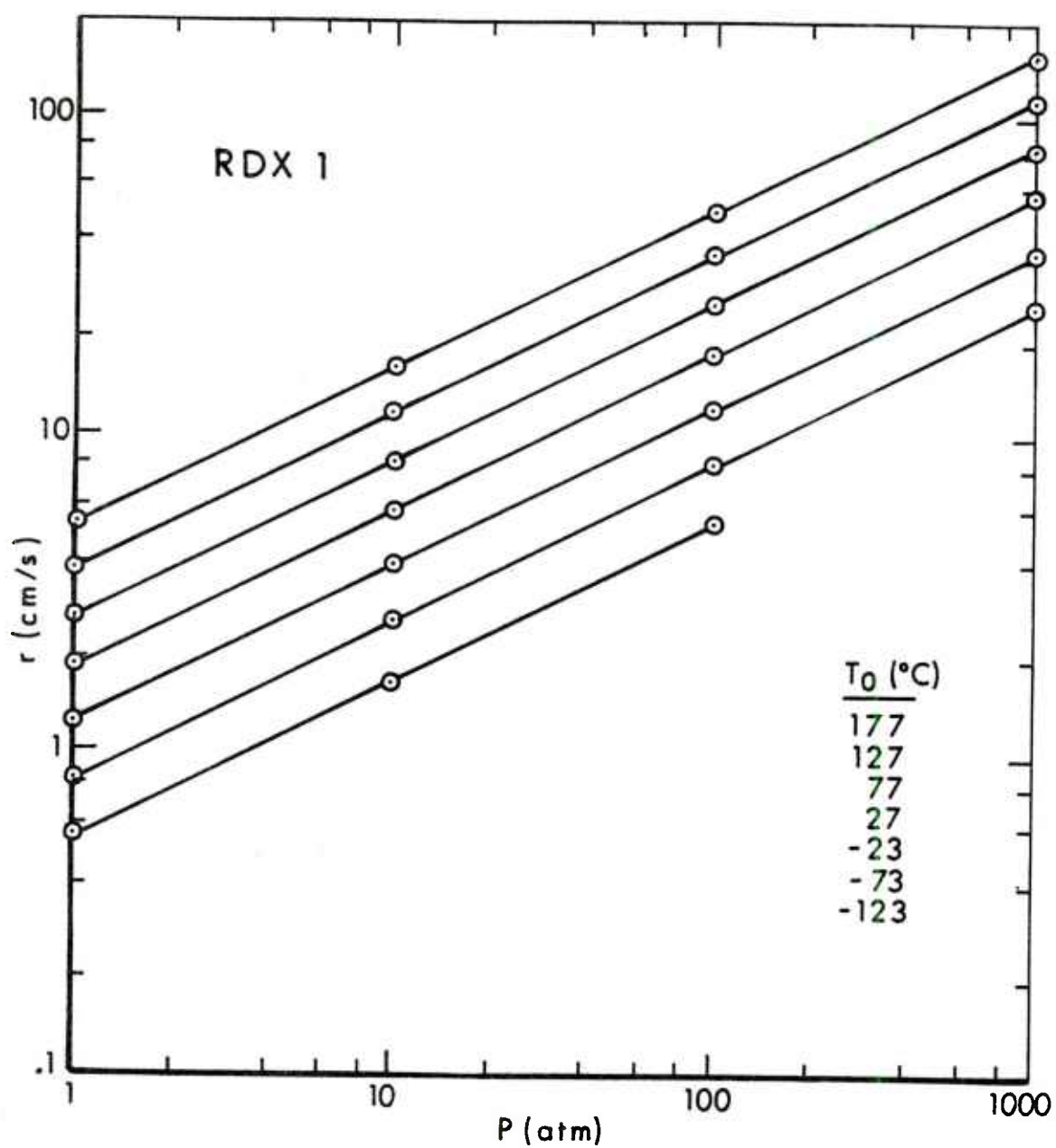


Figure 1. Burning rate vs. pressure computed for the RDX1 data set at various initial temperatures.

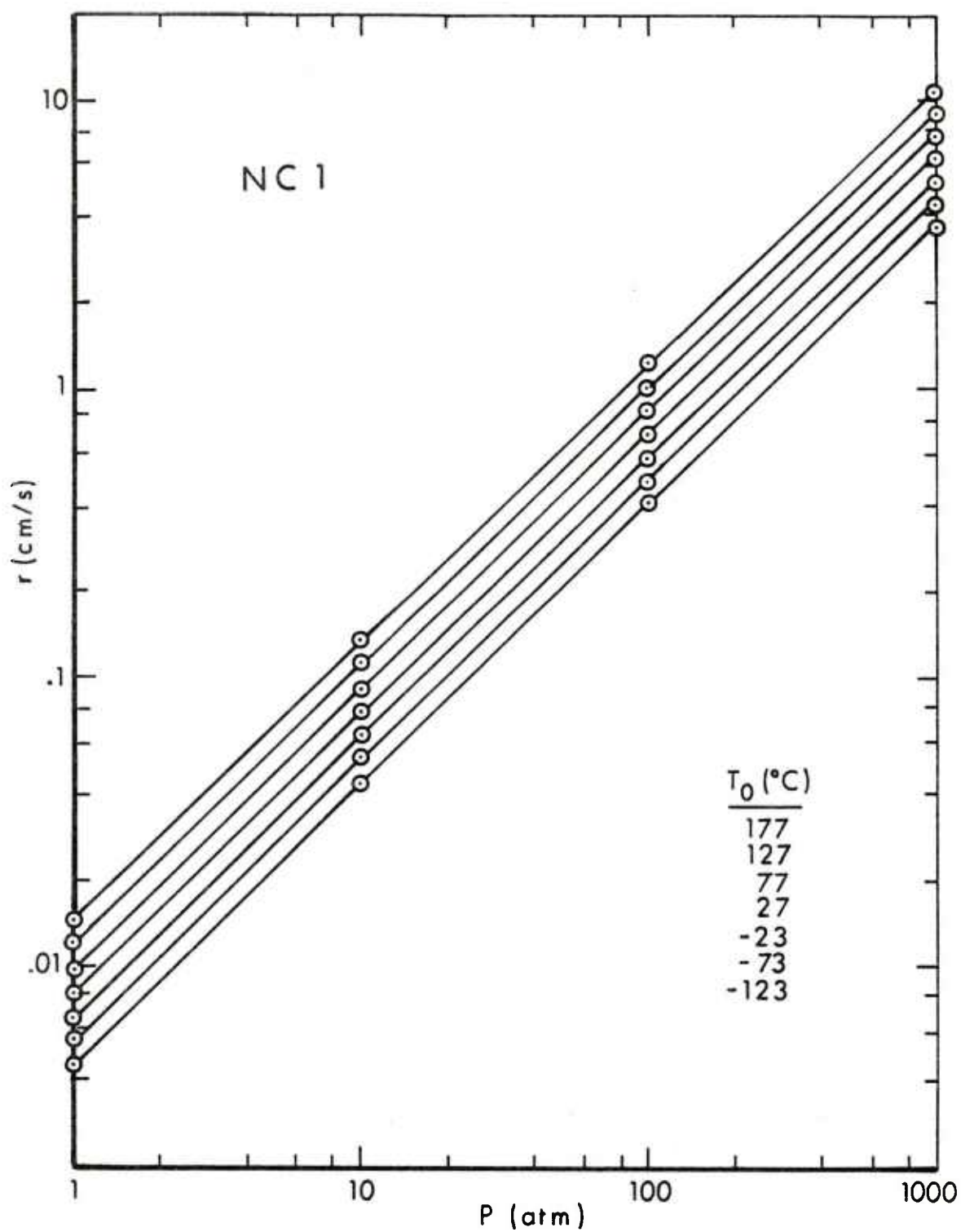


Fig. 2. Burning rate vs. pressure computed for the NC1 data set at various initial temperatures.

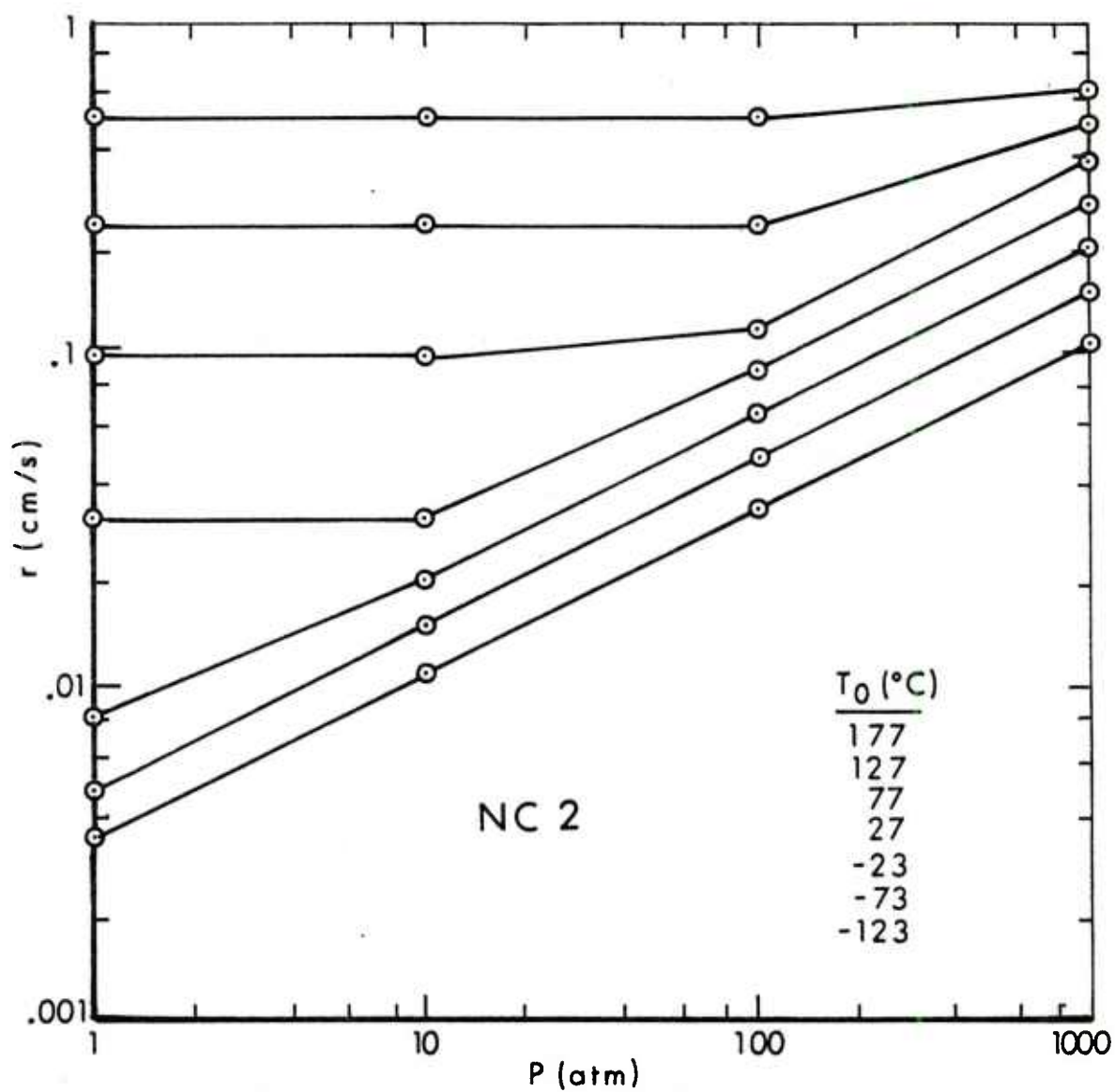


Fig. 3. Burning rate vs. pressure computed for the NC2 data set at various initial temperatures.

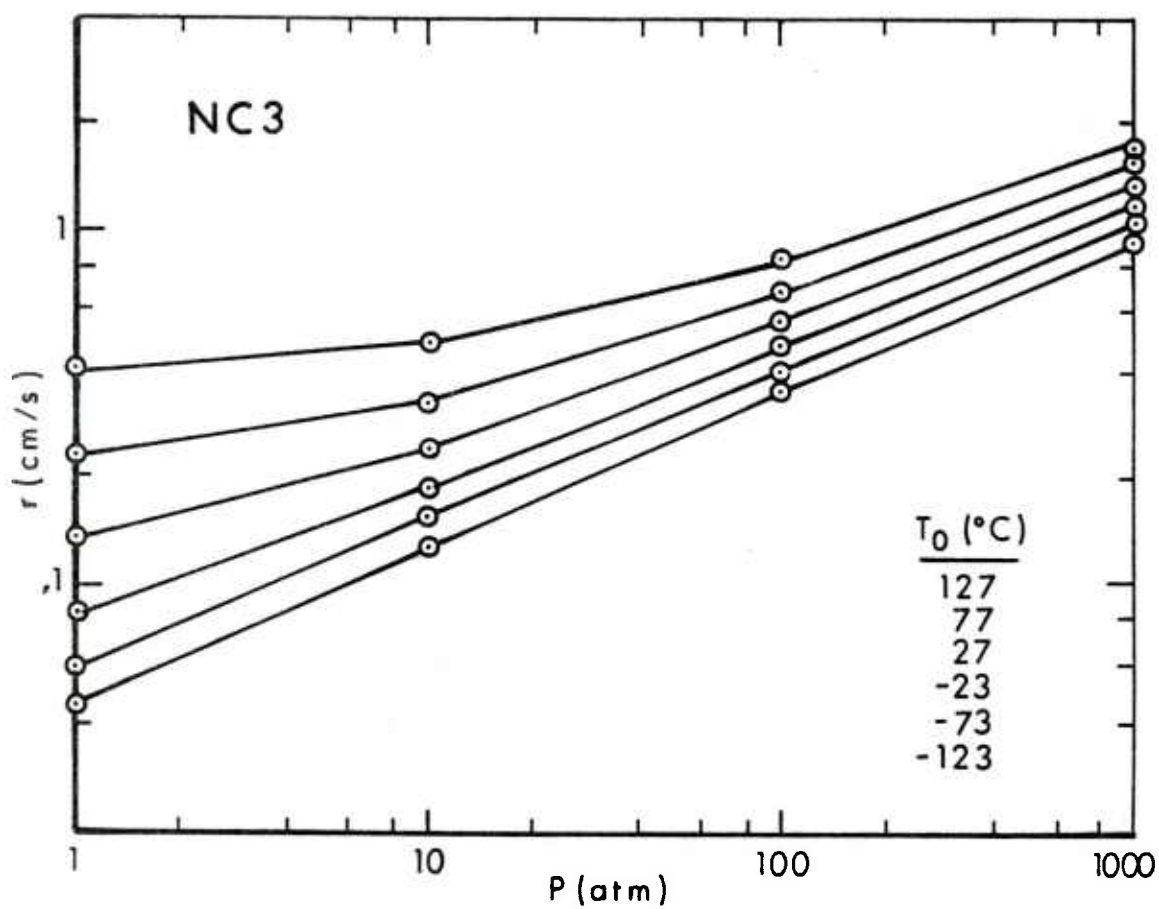


Fig. 4. Burning rate vs. pressure computed for the NC3 data set at various initial temperatures.

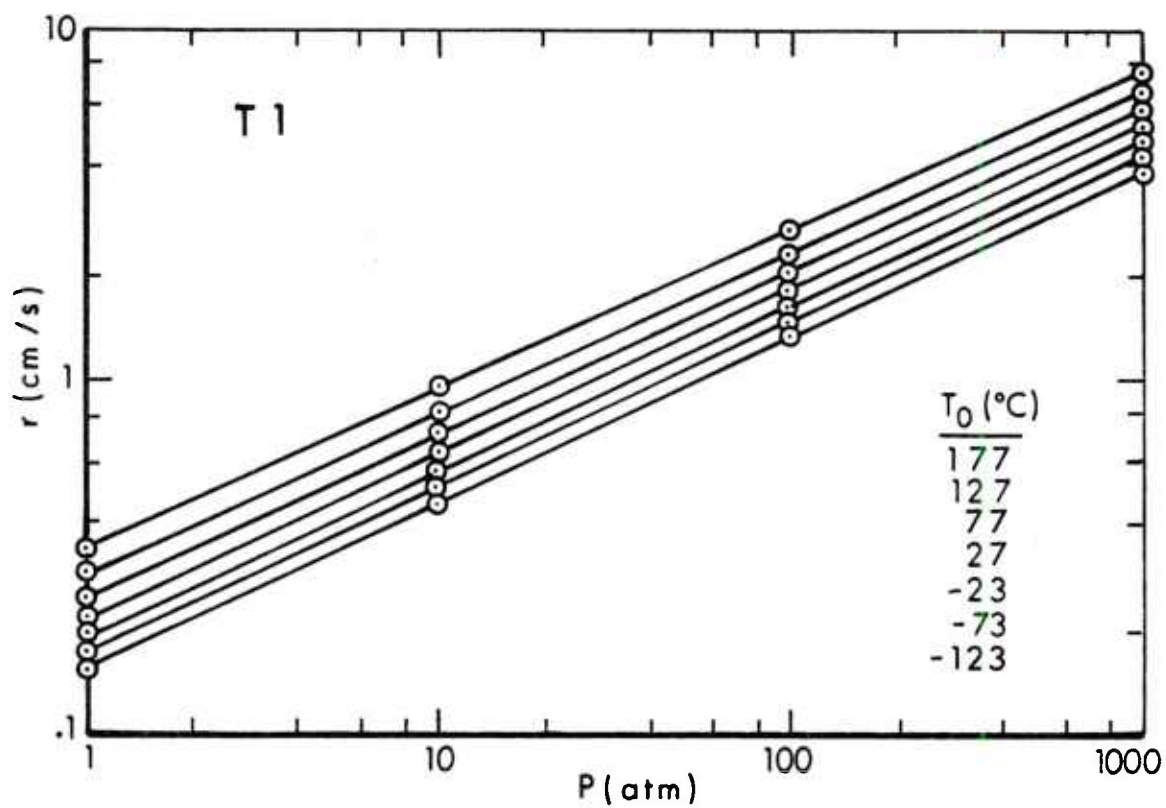


Fig. 5. Burning rate vs. pressure computed for the T1 data set at various initial temperatures.

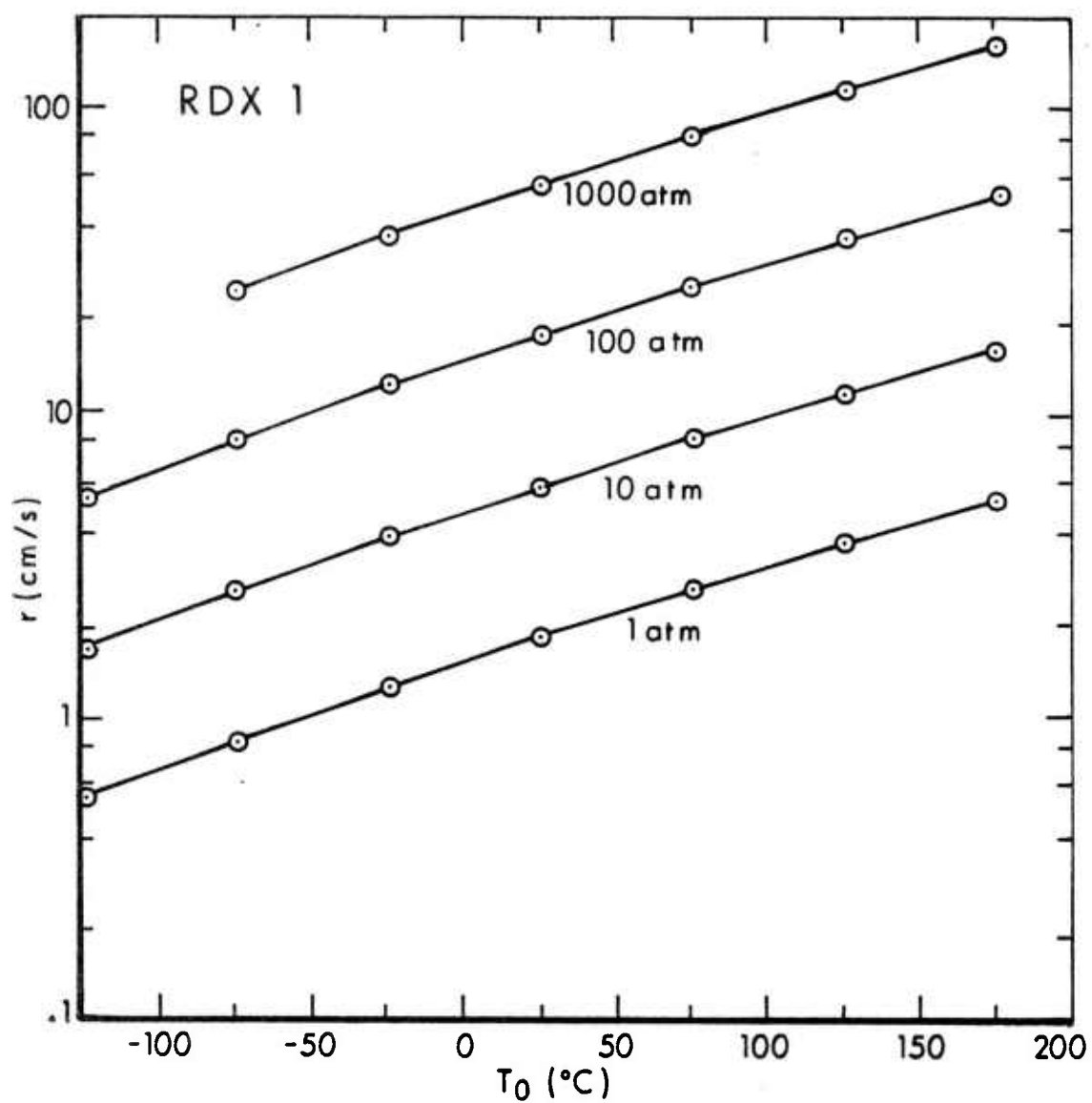


Fig. 6. Burning rate vs. initial temperature computed for the RDX1 data set at various pressures.

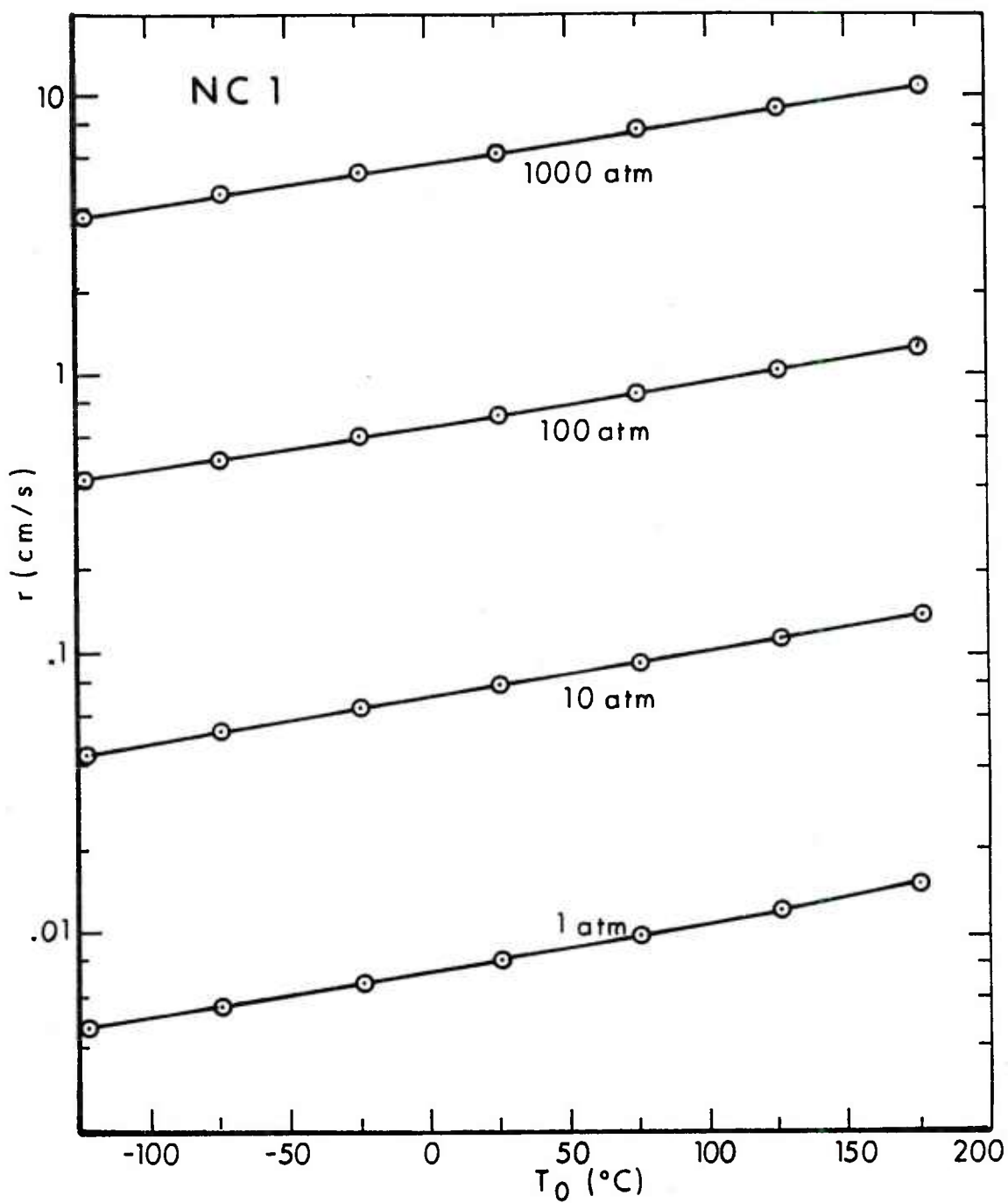


Fig. 7. Burning rate vs. initial temperature computed for the NC1 data set at various pressures.

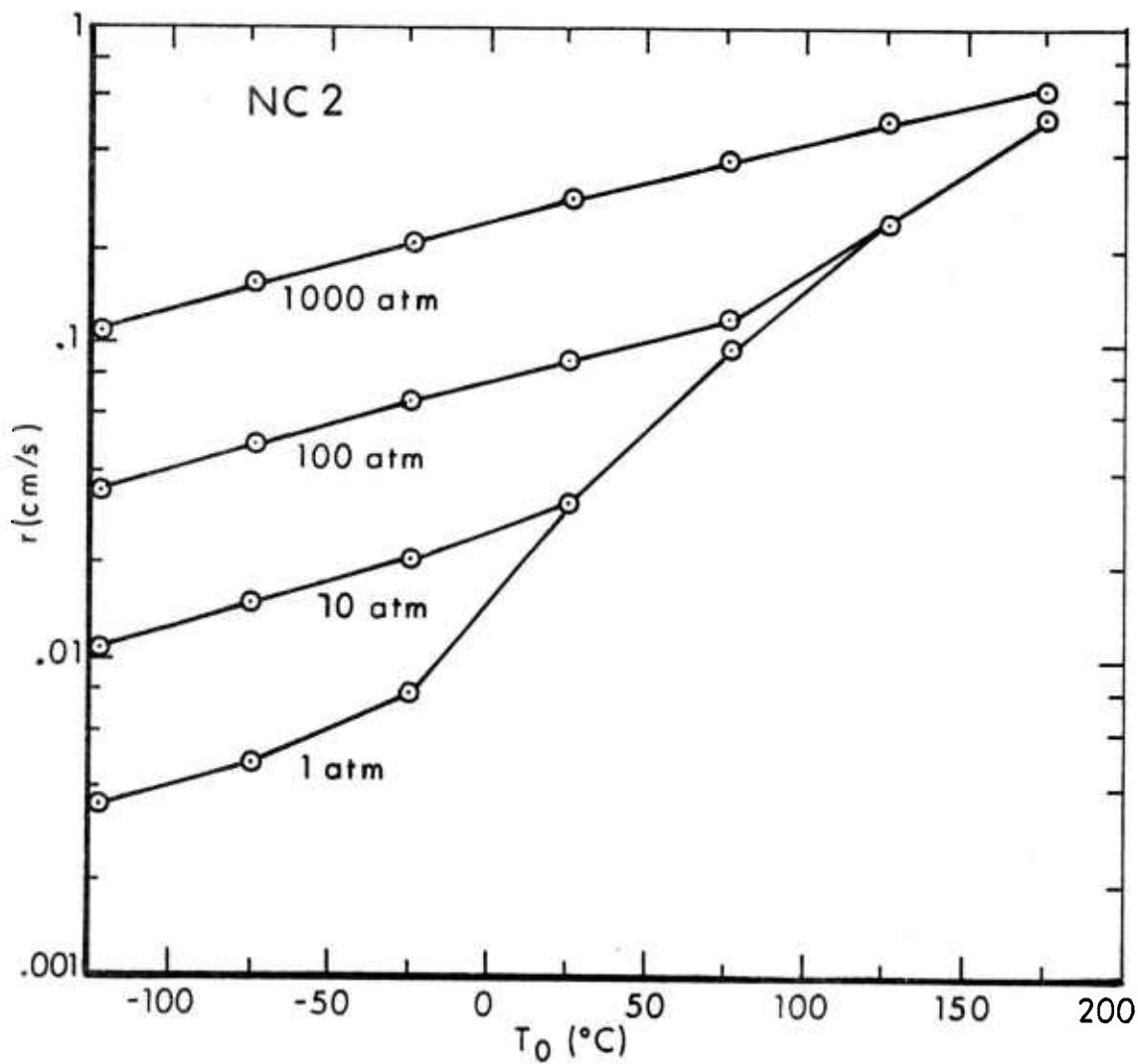


Fig. 8. Burning rate vs. initial temperature computed for the NC2 data set at various pressures.

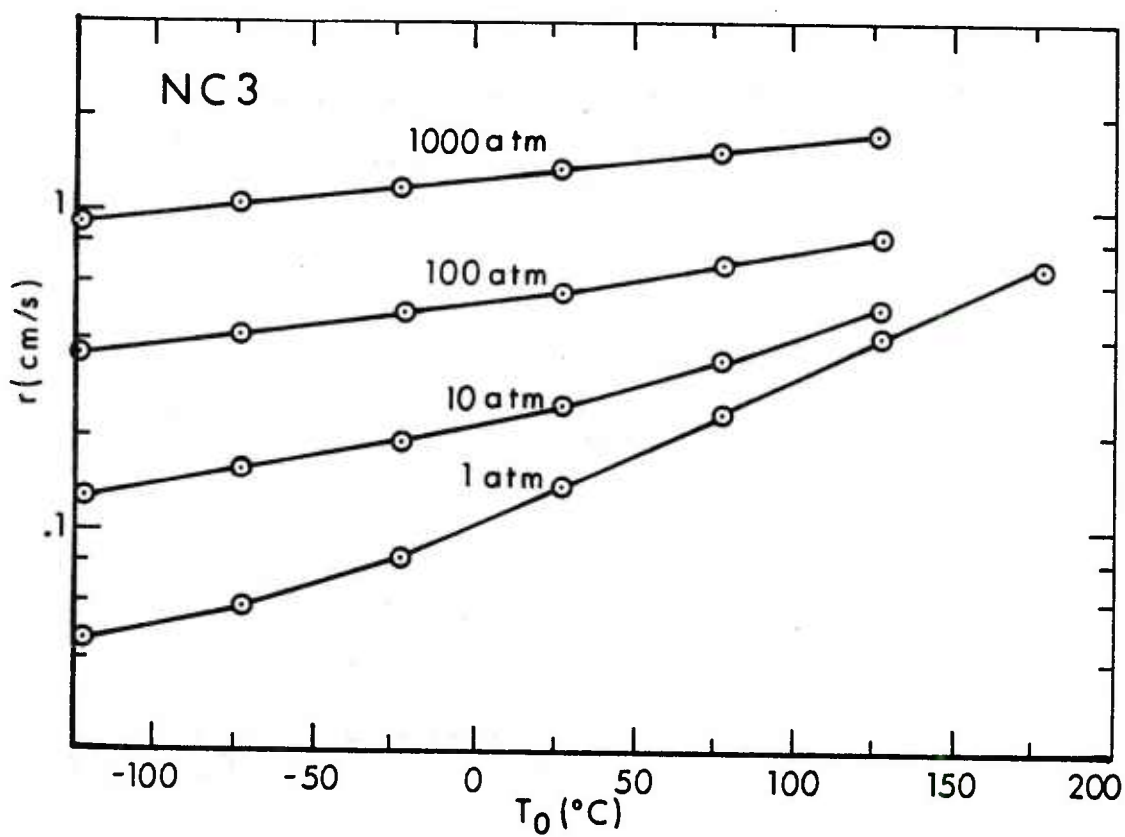


Fig. 9. Burning rate vs. initial temperature computed for the NC3 data set at various pressures.

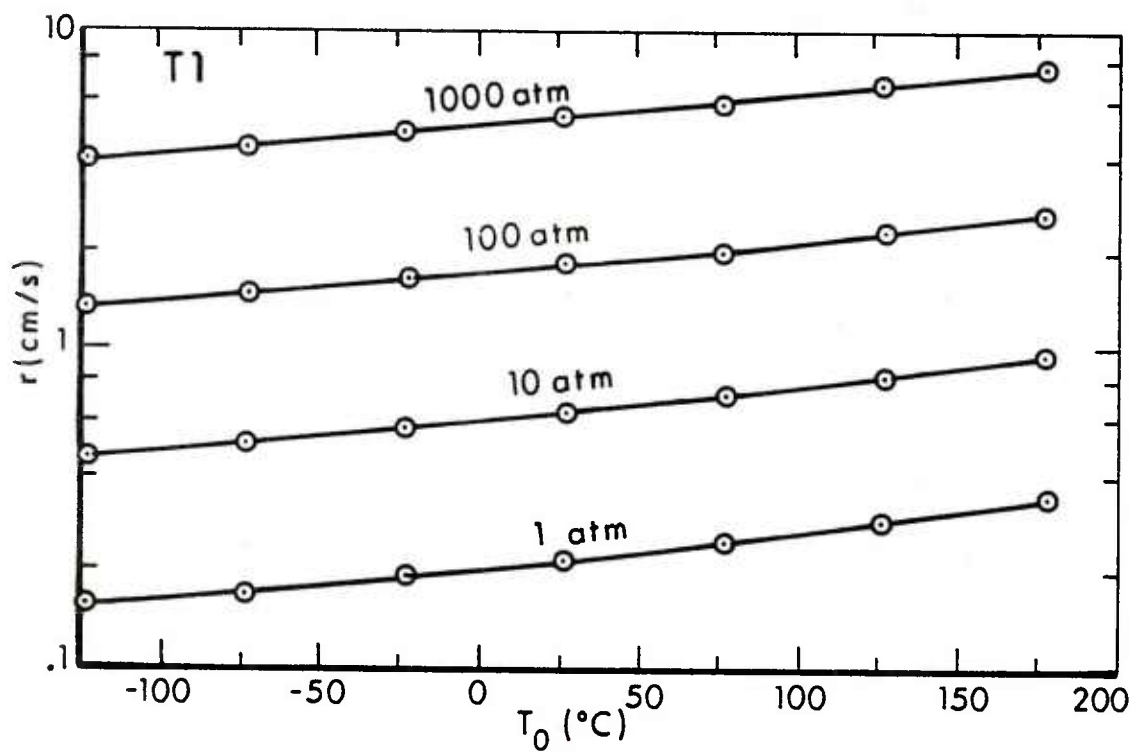


Fig. 10. Burning rate vs. initial temperature computed for the T1 data set at various pressures.

pressure deflagration limit reached when essentially all of the gas heat release is returned to the surface has no practical effect on the model propellant burn rates. Its influence can be detected however in the slight pressure index decrease between 100 and 1000 atm for NC3 at the lowest T_0 . For some other combination of parameters its effect may be greater. Note that for several of the model propellants no departure of the rate from P^n dependence is observed over the chosen 1-1000 atm range even though the value of n does decrease slightly with increasing T_0 .

The initial temperature dependence of the model burning rates is displayed as $\ln r$ vs. T_0 to facilitate comparison with the real propellant burning rates to be discussed. This dependency is usually discussed, however, in terms of the temperature sensitivity, σ_p , defined as the slope of the $\ln r$ vs. T_0 curve. The solid curve in Fig. 11 summarizes the general shape of $\sigma_p(T_0)$ found for the model propellants. At a higher pressure the curve shifts to the right as indicated by the dashed line. ($\sigma_p(T_0)$ is plotted explicitly for each data set in Ref. 2). Any given model propellant will exhibit only part of this characteristic curve over the practical range of T_0 and P . The shape of $\sigma_p(T_0)$ in Fig. 11 arises from the transition between a laminar flame character at low T_0 to a rate which is completely controlled by the heat from the solid reaction^o at high T_0 . Movement on the curve through the transition region from left to right indicates a decrease in the contribution to the surface temperature coming from the gas. This occurs when increases in T_s (due to increases on T_0) result in a lower gas-phase heat feed-back. This happens when the increase in mass flow accompanying the T_s increase overcomes the increased gas reaction, blowing the flame further from the surface.

In order to compare these model propellant trends to real propellants we have collected representative data for pure nitramines ("monopropellants") and nitrate ester propellants in Figs. 12-19. For ready comparison these data are plotted on the same scales used for the model propellants.

As with the model propellants the real propellant burning rates can be well represented by the form $r=a+bP^n$. The maximum pressure index is about .8 - .9 compared to the model values near .5 or 1. Also, like the model propellants, there is a slight decrease in the value of the pressure index at any given pressure as the initial temperature is raised. Taking 20 atm and 25°C as an arbitrary reference, the real burning rates are closely grouped at .3 - .45 cm/sec whereas the model propellants span the wider range .1 - 8 cm/sec.

At 20 atm and 25°C the nitrate ester propellants have a σ_p of about .0056 - .0059°C⁻¹ and the value for HMX is about .0023°C⁻¹. The model propellant σ_p is in the range .0023 - .0076°C⁻¹ at the same temperature and pressure. Over changes of about 50°C little change in σ_p occurs for both real and model propellants. The Zenin data²³ (Fig. 19) cover the widest range of T_0 and show σ_p increasing with increasing T_0 . Other nitrate ester data (Fig. 17 and 18) show a nearly constant σ_p over the measured range.

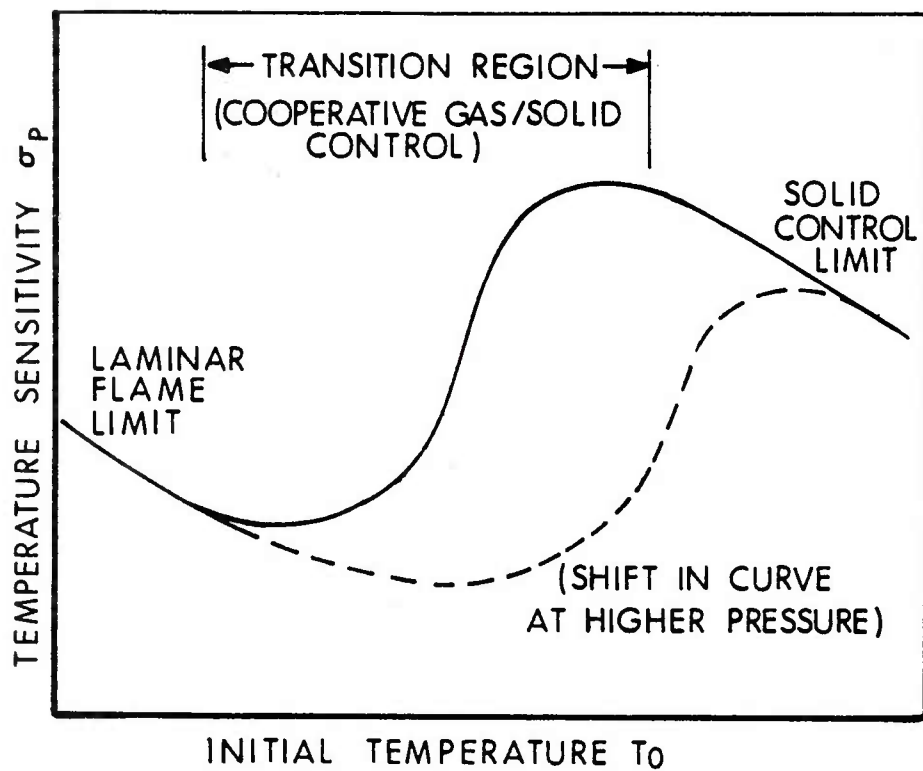


Fig. 11. Patterns of burning rate control evidenced by temperature sensitivity calculations for idealized propellants.

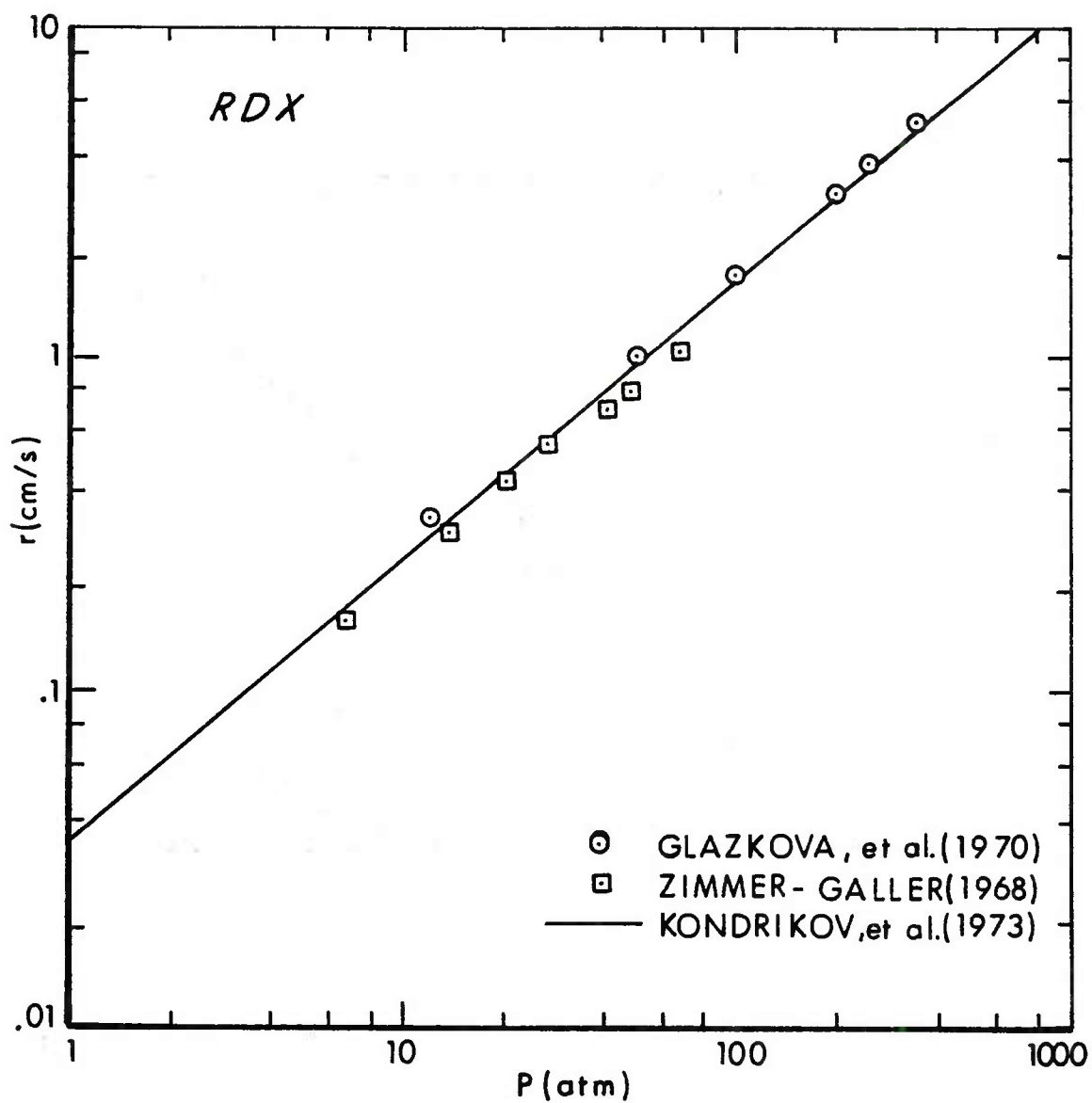


Fig. 12. Reported measurements²⁸⁻³⁰ of burning rate vs. pressure for RDX monopropellant.

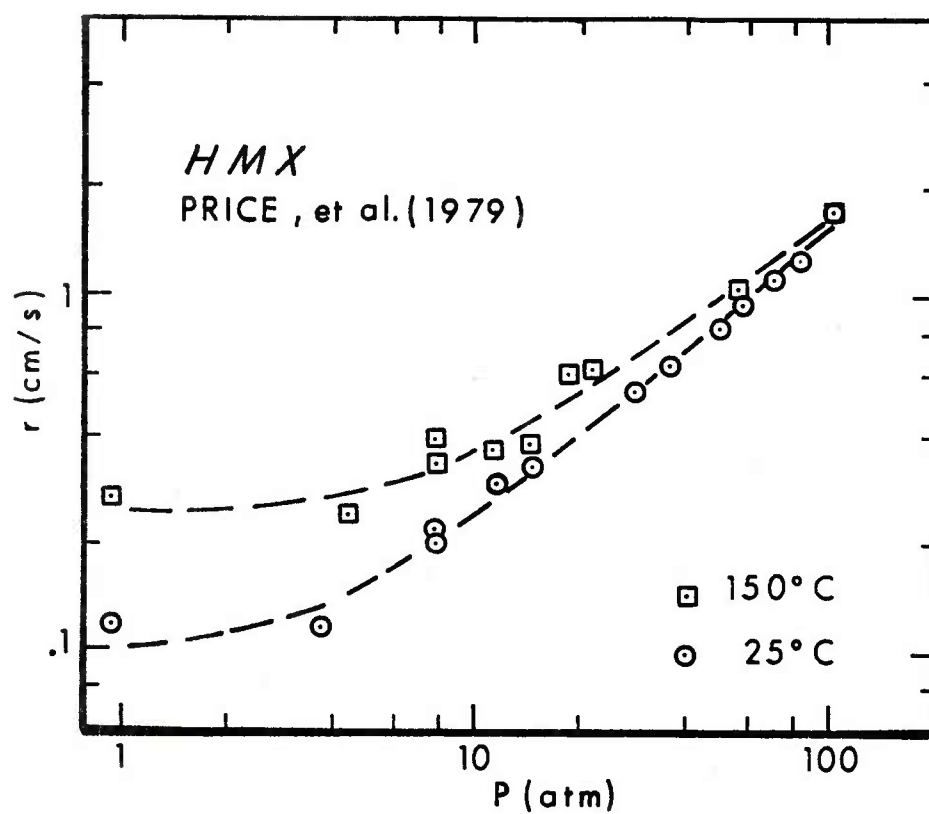


Fig. 13. Reported measurements³¹ of burning rate vs. pressure for HMX monopropellant.

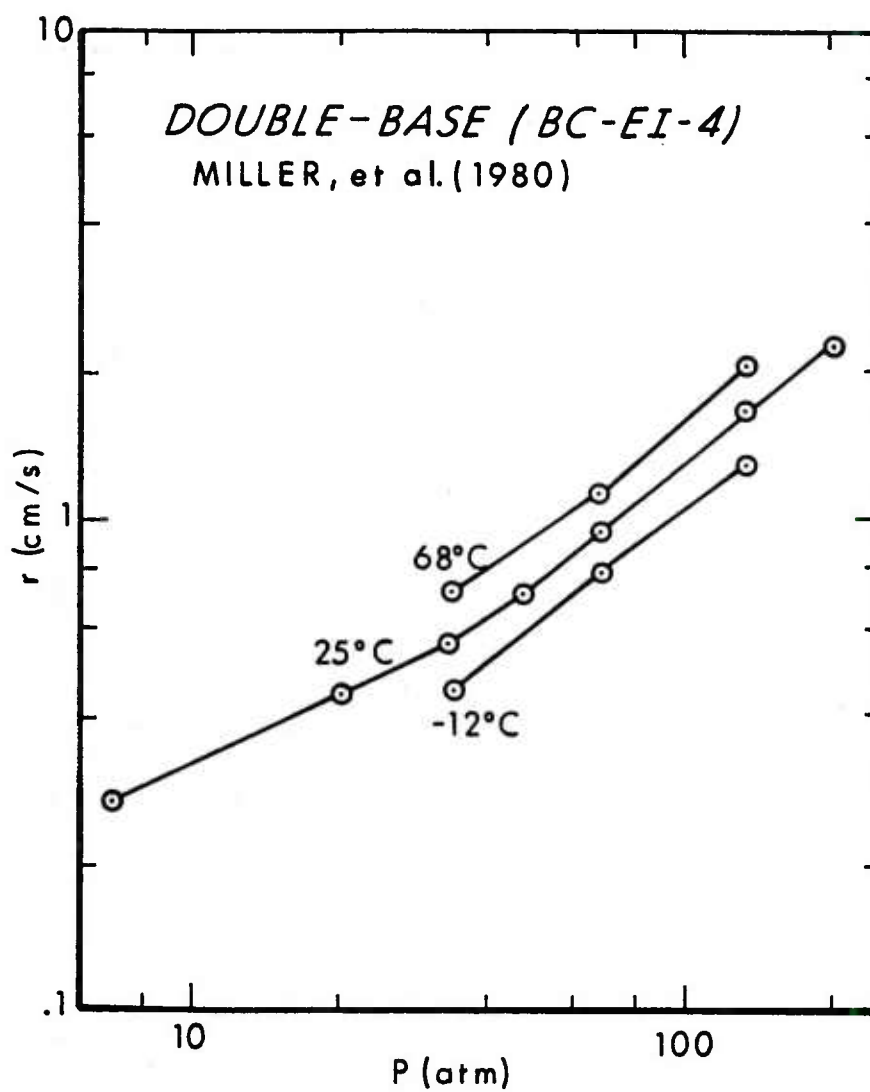


Fig. 14. Reported measurements³² of burning rate vs. pressure for a double-base propellant (coded BC-EI-4 in Ref. 32).

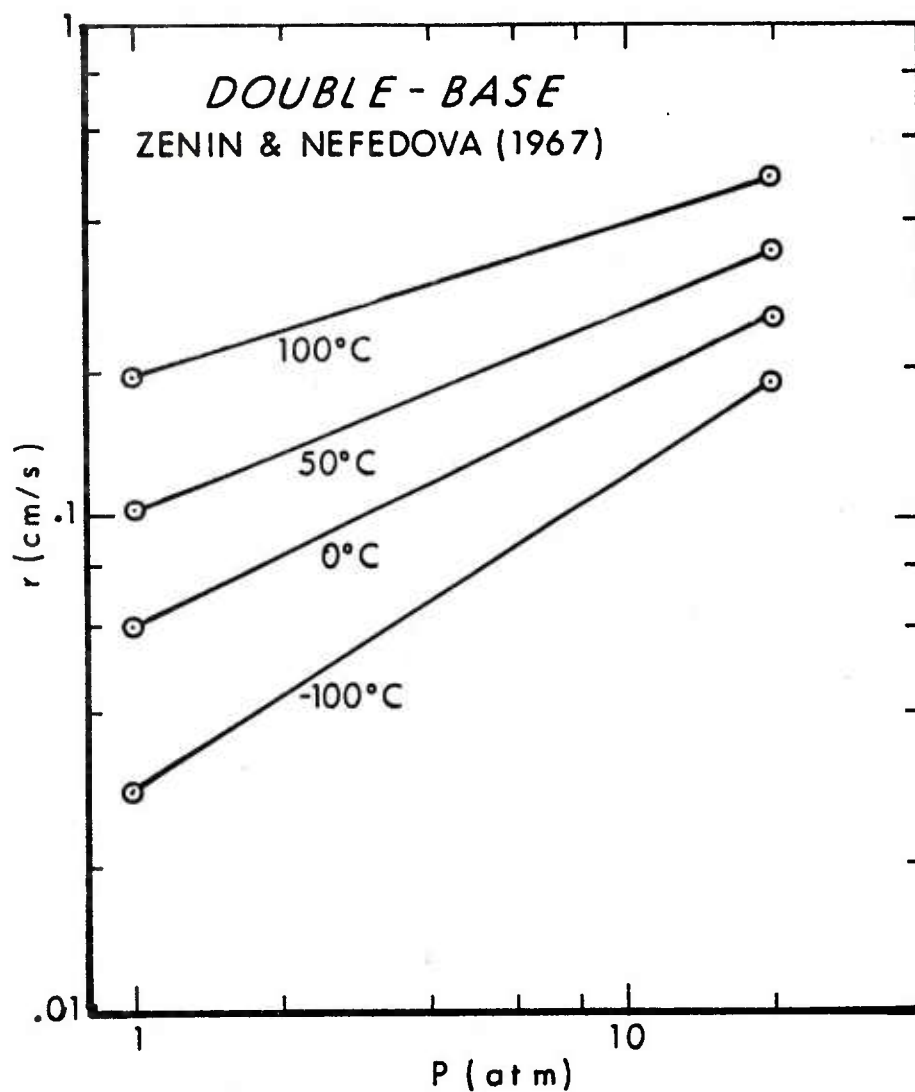


Fig. 15. Reported measurements²³ of burning rate vs. pressure for a double-base propellant (N powder).

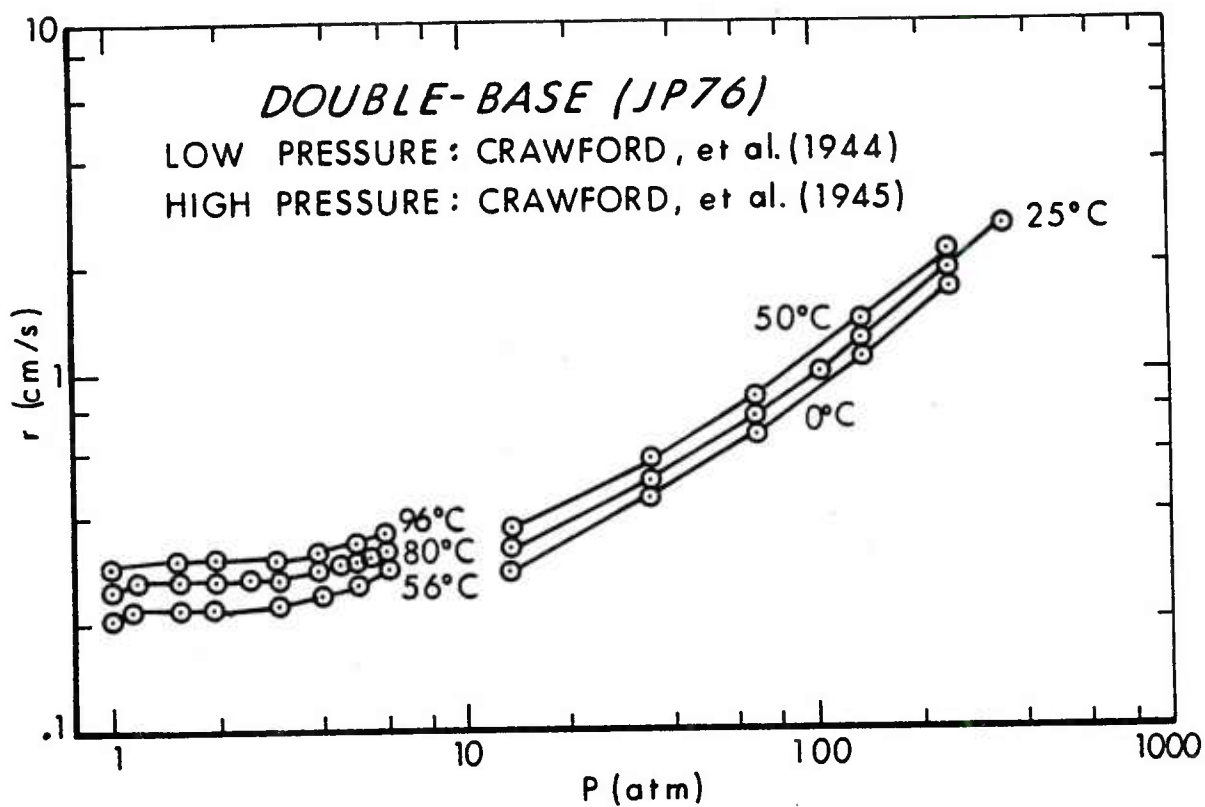


Fig. 16. Reported measurements^{33,34} of burning rate vs. pressure for a double-base propellant (JP76).

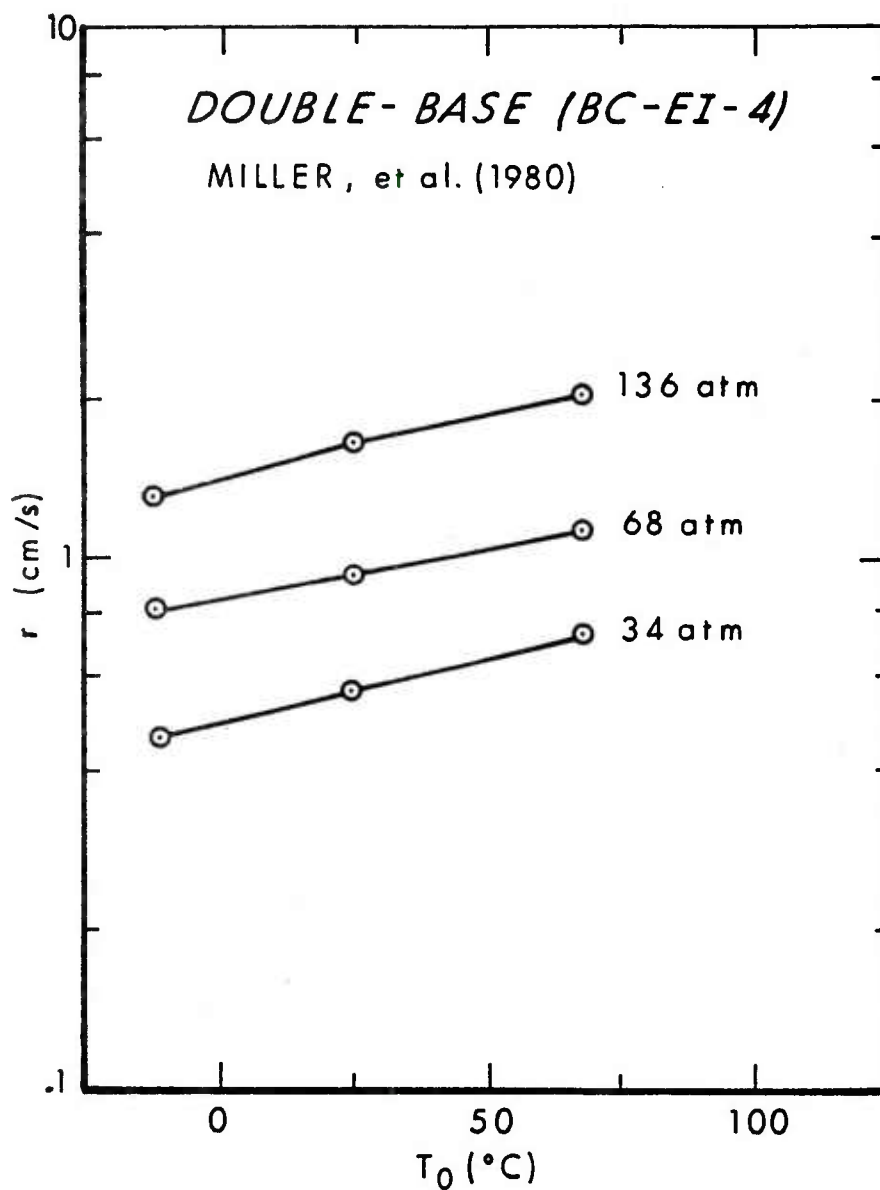


Fig. 17. Reported measurements³² of burning rate vs. initial temperature for a double-base propellant (coded BC-EI-4 in Ref. 32).

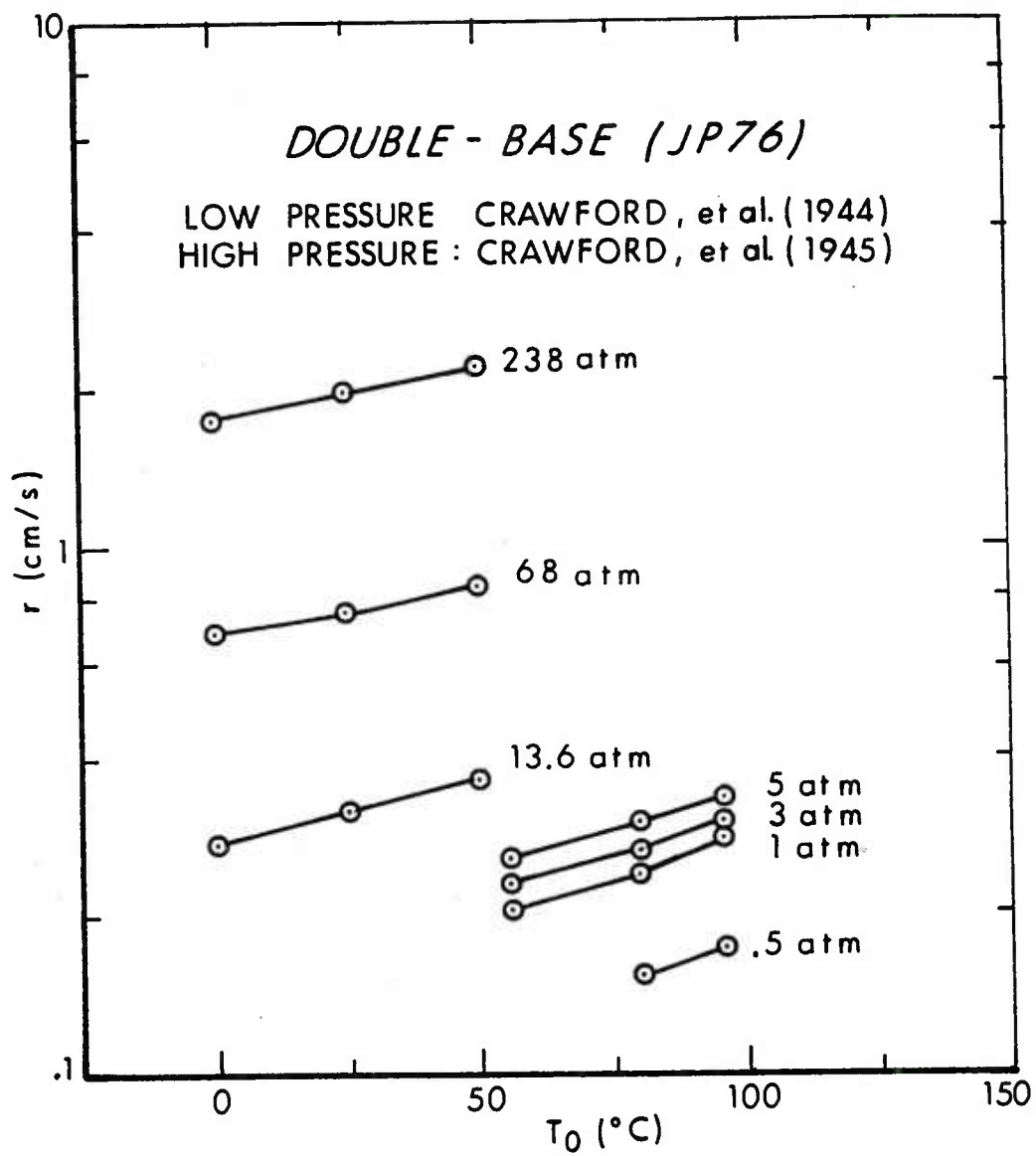


Fig. 18. Reported measurements^{33,34} of burning rate vs. initial temperature for a double-base propellant (JP76).

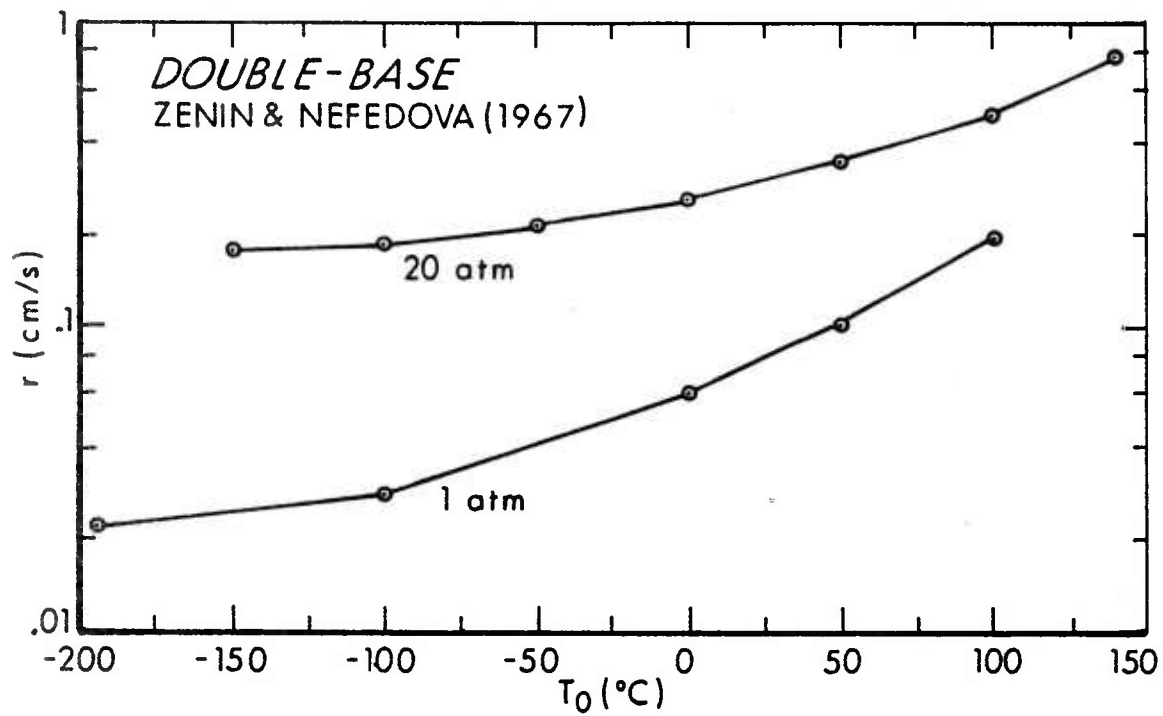


Fig. 19. Reported measurements²³ of burning rate vs. initial temperature for a double-base propellant (N powder).

This comparison of real and model propellant phenomenology can be used to suggest likely weaknesses in the idealization under consideration. The discrepancy in maximum pressure index might arise from several sources. Thermo-couple data^{8,9} have indicated that both the solid phase heat release (Q_s) and the dark zone temperature (T_f) increase with pressure. Either effect could alter the effective pressure index although neither would affect the qualitative behavior of σ_p in Fig. 11. The assumption of constant conductivity and specific heats is also only a rough approximation and could influence the pressure index. The assumption of a single elementary reaction in each phase is only a convenient idealization. The presence of parallel reactions of different orders might cause the pressure index to increase with increasing pressure. It would seem, however, that little would be gained by adding any of these refinements unless they first could be characterized through experiments.

IV. CONCLUSIONS

We have described a systematic study of the phenomenology associated with a typical idealization of solid propellant combustion. Many of the trends identified for the "model propellants" were shown to have parallels in real propellant combustion. The general pressure dependence of the burning rate for both model and real propellants is similar but has too little structure to be of much heuristic value. The temperature sensitivity as a function of initial temperature for various pressures has a richer structure, the details of which correlate with the relative control of the burning rate by solid and gas phase processes. Various experimental σ_p values follow different features of the model propellant pattern, but in most cases data over a wider range of T_0 are needed to confirm the parallels.

The Classical Approach to propellant modeling, if properly applied, can be a useful method of exploring the cooperative effects of proposed combustion mechanisms. An important limitation to its use, however, derives from the fact that errors in the mechanism description often have a weak effect on the burning rate. An erroneously high reaction rate in the gas phase, for example, tends to increase the heat feedback which leads to a higher burning rate. But the magnitude of the burning rate increase is moderated by an increased convective heat loss. The net effect is that the burning rate may not be a sensitive function of the mechanisms on which it depends. For this reason the next step in modeling propellant combustion, in our opinion, will have to be based on mechanisms independently validated for each phase. Such mechanisms need not be greatly increased in sophistication or chemical specificity but must be based on characterizations of the solid and gas phase processes which are consistent with experimental measurements of the detailed combustion wave structure.

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LIST OF SYMBOLS

- A_G - gas-phase reaction frequency factor
- A_S - condensed-phase reaction frequency factor
- B - gas-phase reactant label
- C_p - specific heat for solid and gas phases
- E_G - activation energy for gas-phase reaction
- E_S - activation energy for solid-phase reaction
- M - mass regression rate (mass flux)
- M_O - constant in pyrolysis surface decomposition mechanism
- m_B^{-0}, m_B^{+0} - mass fraction of B evaluated at negative and positive sides of the solid/gas interface, respectively.
- N_O - Avogadro's number
- N_2 - number of moles of C produced per mole of B which reacts
- n - pressure exponent of burning rate
- P - total pressure
- Q_S - heat of reaction per unit mass for solid reaction (positive for exothermic)
- Q_G - exothermic gas-phase reaction heat per unit mass of B
- R - universal gas constant
- r - linear regression rate of propellant surface
- T - local temperature
- T_O, T_S, T_f - initial, surface, and flame temperatures, respectively
- W_A, W_B, W_C - molecular weights of A, B, and C
- \bar{W} - average molecular weight of mixture in gas-phase
- λ - heat conductivity in gas-phase
- ν - gas-phase reaction order
- ρ_S - mass density of solid
- σ_p - temperature sensitivity at constant pressure

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